# Preface to Special Topic: Selected Papers from the Piezoresponse Force Microscopy Workshop Series: Part of the Joint ISAF-ECAPD-PFM 2012 Conference

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# Preface to Special Topic: Selected Papers from the Piezoresponse Force Microscopy Workshop Series: Part of the Joint ISAF-ECAPD-PFM 2012 Conference

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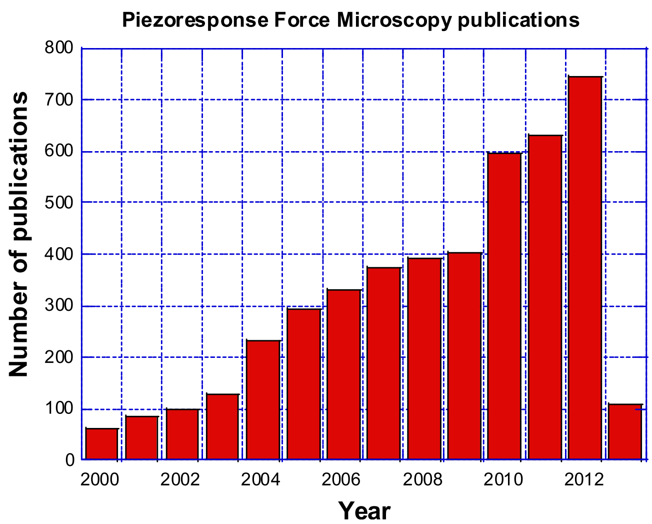
We are pleased to present the fourth edition of the Special Topic Issue on Piezoresponse Force Microscopy and Nanoscale Phenomena in Polar Materials published as a part of the joint IEEE ISAF-ECAPD Conference. In the last two decades since its inception,[1–6] piezoresponse force microscopy (PFM) has emerged as an indispensable tool for characterizing and manipulating piezoelectric and ferroelectric materials and structures at the nanoscale,[7–9] and the field has witnessed rapid growth since the first Special Topic Issue of Journal of Applied Physics on the topic published in 2010.[10] According to Google Scholar, 4479 papers involving PFM have been published, with 2079 of them appeared since 2010 and 852 of them since 2012. Rapid growth of the PFM papers is illustrated in the following graph where a cusp on the curve is related to the above mentioned 2010 Issue. This special issue is a timely collection of recent advances in PFM techniques, theories, and applications.




Indeed, in the last three years or so, multiple breakthroughs related to PFM have been made, and many of the advances have been envisioned in the Preface to the first special issue.[10] For example, the feasibility of local electromechanical measurements for mapping electrochemical reactions has been suggested,[11] which has since been realized, resulting in a new electrochemical strain microscopy (ESM) technique that has been implemented in commercial systems.[12] The ESM is now being practiced by a number of groups in studying electrochemical systems, including lithium ion batteries,[13–17] solid oxide fuel cells,[18] metal-air batteries,[19,20] and supercapacitors.[21] In the present issue, Karapetian and Kalinin developed a timely theoretical analysis of indentation of materials with chemical species distribution, which can be used to analyze and interpret imaging mechanism of ESM.[58] Furthermore, the 2010 Preface also predicted opportunities for new discoveries and breakthroughs in biological systems and strongly correlated oxides, and one of the exciting developments in the last two years is the observation of biological ferroelectricity in seashells,[22] aortic walls,[23] elastin,[24] glycine,[25] and peptide nanotubes,[26] which came more than 50 years after the closely related piezoelectricity was reported in biological tissues and 5 years after ubiquitous presence of biological piezoelectricity was established by PFM.[27–31] This advance is undoubtedly enabled by PFM, as vividly illustrated by Li and Zeng in their detailed studies on electromechanical coupling and ferroelectric switching of seashell in the present issue.[59] Another exciting development is recent realization of polarization reversal by mechanical stress,[32] attributed to the flexoelectric effect afforded by nanoscale Scanning Probe Microscopy (SPM) tip. A couple of studies in the present issue devote to effects of mechanical field as well. Morozovska *et al.* studied domain wall interactions with elastic defects,[60] and Xu and Kan *et al.* investigated the effects of stress on ferroelectric aging,[61] linking it to the intrinsic mechanisms of domain wall mobility. These selected examples highlight the vibrant development of the field in recent years.

The present special issue of Journal of Applied Physics contains selected papers from the International Symposium of Piezoresponse Force Microscopy (PFM) and Nanoscale Phenomena in Polar Materials held in Aveiro, Portugal on July 9 13, 2012, in conjunction with the 21st IEEE International Symposium on Applications of Ferroelectrics (ISAF) and 11th European Conference on the Applications of Polar Dielectrics (ECAPD). The symposium featured a set of invited and contributed talks on principles and applications of PFM and continued the series of Piezoresponse Force Microscopy Conferences and Workshops started in 2007 in Oak Ridge (USA) and continued in Lausanne (Switzerland) and Oak Ridge (USA) in 2008 and 2013, Aveiro (Portugal), Tsukuba (Japan), and Montreal (Canada) in 2009, Beijing (China) and Prague (Czech Republic) in 2010, and Vancouver (Canada) in 2011. These workshops comprise tutorials on fundamentals of PFM operation, relevant instrumental details, and image and electromechanical hysteresis interpretation, experimental hands-on classes in the PFM laboratories equipped with modern PFM setups, as well as latest development in the field, with the selected papers presented in this Special Topic Issue.

A variety of ferroelectric materials have been investigated in the present issue, including piezoelectric transducer lead zirconium titanate (PZT) thick films synthesized from composite sol gel,[62] thickness dependence of structure and piezoelectric properties of polycrystalline thin films.[63–65] In particular, Liu *et al*.[62] have demonstrated that the PZT and lead magnesium niobate-lead titanate (PMN-PT) thick films prepared by modified sol-gel method possess local piezocoefficients comparable to those of their bulk counterparts. A number of papers devoted to PFM study of ferroelectric materials under thermal field or electrochemical process. In their contribution,[66] Jankowska-Sumara *et al*. detail intricate set of temperature- and field-induced phenomena in the relaxor ferroelectrics as accessed by variable temperature PFM. Guo *et al*.[67] report the detailed studies of temperature-induced evolution of structure and electromechanical response in ferroelectric polymer, and Shur *et al*.[68] and Rodrigiez *et al*. communicate their findings of light-induced photochemical reactivity on the ferroelectric $LiNbO_3$ surfaces,[69] providing new insight into physics and electrochemistry of ferroelectric-semiconductors.[33–35] The role of surface humidity of surface potential was explored by Verdaguer,[70] complementing earlier data on $BaTiO_3$ surfaces.[36–40] Several studies combine PFM and other localized characterizations, such as atomic force acoustic microscopy (AFAM)[41,42] by Zhou and Li *et al*.,[71] and confocal Raman microscopy[72] and Raman spectroscopy.[73] In particular, combination of PFM and AFAM allows direct insight into the coupling between mechanical and ferroelectric properties, and hence fundamental mechanics of ferroelectric materials[43–49] Finally, the review by Barrett *et al*.[74] reports systematic studies of polarization controlled surface electronic properties and domain structures on ferroelectric surfaces using a variety of electron imaging and structural probes. These studies provide much needed insight into the electronic properties of ferroelectric surfaces, complementing recent advances in X-ray scattering in reactive environment achieved by Argonne group.[50–53]

Multiferroic materials continue to be a hot topic. Using PFM, Karpinsky *et al*. investigated $BiFeO_3$-$LaFeO_3$-$CaTiO_3$ ceramics near the rhombohedral-orthorhombic phase boundary and observed gradual decrease of piezoelectric response in $BiFeO_3$-$LaFeO_3$-$CaTiO_3$ system with CaITi content increase, except at a narrow concentration region near polar-antipolar phase boundary where piezoelectric signal shows maximum value.[75] Venkata Ramana *et al*. confirmed that Fe-doped $BaTiO_3$ thin films have a room-temperature ferromagnetism and retain switchable ferroelectric properties up to 10 mol. % of Fe.[76] In another study, Domingo *et al*. have observed two different layers on the surface of high-quality $BiFeO_3$ single crystals. One layer is just a dead layer of the thickness of 5 nm and another one is characterized by the presence on nanotwins that is very important for predicted photovoltaic and spin-dependent properties.[77] Lazarević *et al*. reported synthesis of nanodimensional spinel $NiFe_2O_4$ and $ZnFe_2O_4$,[78] and these magnetic nanomaterials can benefit from recently developed piezomagnetic force microscopy (PmFM),[54] which detect magnetic field induced surface vibration due to magnetostrictive effect, similar to PFM.

The analyses and interpretations of PFM and related techniques have received continued attention. Theoretical model of SPM tip electrostatic field has been developed, effective piezoelectric response of twin walls has been analyzed,[79] and resolving ferroelectric nanostructures by PFM has been attempted.[80] In particular, Morozovska *et al*. have calculated the effective piezoelectric response of twin domain walls using decoupling approximation and Landau-Ginzburg-Devonshire theory. The strong dependence of the piezocoefficients on the polarization gradient was predicted.[79] The exact solution to the coupled problem of indentation of the punch, subjected to either heat or chemical substance distribution at its base, into three-dimensional semi-infinite transversely isotropic material is presented,[58] which can be used to understand the image formation mechanisms in techniques such as thermal SPM and ESM. In their contribution, Pan *et al*. numerically studied PFM resolution for different kinds of domain walls, in particular, tilted domain wall. They showed that ferroelectric features smaller than the probe diameter could indeed be resolved. Probing ferroelectric structures beneath the surface was also demonstrated.[80]

In the years to come, PFM will continue to play major role in studying ferroelectric, multiferroic, and biological materials and systems, enabled by advanced imaging and spectroscopy techniques, *in-situ* observation and manipulation in combination with other microscopic tools, rigorous mathematical analysis and phase field simulations, as well as PFM in non-ambient and more challenging environment, such as high temperature, high vacuum, and in liquid. Exciting opportunity exists in biology, and we expect ferroelectricity to be found in many more biological materials and structures and believe PFM will help uncover its molecular mechanisms and physiological significance. For example, a recent PFM study reveals that ferroelectric switching of elastin is suppressed by glucose,[24] which could be related to glycation and aging. Great opportunity also exists in molecular and organic ferroelectrics, which starts to show properties comparable to perovskite oxide recently,[55–57] and the low symmetric molecular crystal offers much richer phenomena for PFM to probe. Other dynamic strain based SPM techniques similar to PFM will continue to flourish, particularly the ESM for energy materials and systems, and magnetostrictive based PmFM will also emerge as a powerful tool to probe magnetic materials and structures at the nanoscale.

We are extremely grateful to all the attendees of the IEEE ISAF-ECAPD-PFM 2012 joint conference that gathered a record number of participants. The increase attendance and above mentioned rapid growth of the PFM publications are both a sign of the tremendous success of this field. We thank all contributors for their significant commitment to disseminate the results of their work in this Special Topic issue.

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